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Contribution of neutron-capture reactions to observed tungsten isotopic ratios

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Abstract

To study whether tungsten isotopic ratios observed in terrestrial samples relative to W ratios in an iron meteorite could be made by neutron-capture reactions in the iron meteorite, the fluxes of thermal and epithermal neutrons in a large iron meteoroid, Toluca, were calculated using Monte Carlo particle production and transport codes. Peak neutron-capture rates on W isotopes were calculated to occur at depths of 30–34 cm. The calculated changes in the $^{182}W/^{183}W$ due to neutrons are much less than those observed in terrestrial samples relative to iron meteorites, indicating a non-cosmogenic origin of the observed W isotopic ratios. © 1997 Elsevier Science B.V.

Keywords: tungsten; iron meteorites; isotope ratios; neutrons

1. Introduction

A number of important questions relating to the formation and evolution of the early Earth are still largely unresolved. It was proposed [1] that time constraints on the early development of the Earth, in particular core formation, could be obtained with the use of extinct nuclides, such as the ^{182}Hf–^{182}W system. ^{182}Hf decays to ^{182}W with a half life of 9×10^6 years, which provides a good timescale for studies of both star-forming and planet-building processes. Careful investigation of the ^{182}Hf–^{182}W system is also important in astrophysics because a determination of the initial abundance of ^{182}Hf will provide a key constraint on models of molecular cloud environment within which the Sun formed [2]. Also, several geochemical aspects make the ^{182}Hf–^{182}W chronometer well tailored for dating planetary accretion-related processes, such as core formation [3]. Comprehensive discussions of this system and its advantages can be found in [1,4,5].

High-precision tungsten isotopic analysis indicate the $^{182}W/^{183}W$ ratio in the Toluca iron meteorite is decreased by $(3.9 \pm 1.0) \times 10^{-4}$ relative to a terrestrial standard [6]. Possible causes of this shift are neutron-capture reactions on tungsten isotopes, during Toluca's about 600 Ma exposure to cosmic-ray particles, or radiogenic growth of ^{182}W from ^{182}Hf, either in an undifferentiated chondritic 'reservoir' or in the silicate portion of the Earth after removal of W to the Earth's core, or both, depending on the (at present uncertain) initial abundance of ^{182}Hf [1,4,5].

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Calculations for the rates of neutron-capture reactions on W isotopes were done to study the first possibility. In this paper, we present the results of these calculations and the resulting changes in the ratios of various tungsten isotopes. Some of these results were presented in an earlier conference abstract [7].

2. Calculation model

In our model, the fluxes of particles including thermal and epithermal neutrons, which can readily be captured by tungsten isotopes, are calculated using LCS, the Los Alamos High Energy Transport (LAHET) Code System [8]. LCS is a general purpose, three-dimensional geometry, off-line-coupled, Monte Carlo computer code system that treats the relevant physical processes of particle production and transport. Individual codes in the system treat all interactions of the particles considered within a specified energy range, and particles with energies outside this range are stored in 'history' files, which are used as source of input files for other LCS modules. The high-energy part of neutron interactions and all interactions of heavy ions, protons, pi mesons, muons and other elementary particles are simulated by the LAHET, while neutrons with energies below a cut-off value are written, together with their kinematic parameters, into a neutron file, which is the input file for Monte Carlo N-Particle (MCNP) code [9]. This code transports neutrons down to thermal energies. LCS, its tests, the basics of the built-in physical model, and adaptation to planetary applications are described elsewhere [10] and therefore only the information most important for the present calculations is given here. LCS has been successfully used for studies of spallogenic reactions in stony meteorites [11] and iron meteorites [12]. LCS has also been used to calculate $^{35}Cl(n,\gamma)$ reactions in the 45 cm Knyahinya L5 chondrite [11], reactions involving thermal and epithermal neutrons (production of ^{60}Co, ^{41}Ca and ^{235}U fission rate) in lunar surface [13,14] and for (n,γ) reactions in Mars [15].

The primary cosmic-ray flux at a meteoroid's orbit has two components: galactic and solar. As solar cosmic ray (SCR) particles have low energies, make very few secondary particles, and mainly stop in the outermost few centimetres of the meteoroid, which are usually removed during the passage of the meteoroid through the atmosphere, we consider only galactic cosmic ray (GCR) particles. The GCR particles are a mixture of energetic protons ($\approx 87\%$), alpha particles ($\approx 12\%$), and some heavier nuclei ($\sim 1\%$). The spectral distributions of heavier particles are quite similar to the distribution of protons if energy is taken per nucleon. The GCR proton spectrum was used as input for the LAHET code, which tracks secondary protons and neutrons from intra- and internuclear cascade and evaporation. Proton histories are followed down to an energy of 1 MeV, whereas neutrons below 15 MeV are stored in a history file for further transport with MCNP. The cut-off energy 15 MeV was chosen because the parameters used by LAHET are global averages for all nuclei that become questionable for some nuclei toward lower energies (mainly neutron elastic cross sections) and because individual cross-section libraries for neutron production and transport used by MCNP become sparse toward higher energies. The contributions of alpha (and heavier) particles were simply included in the final results by multiplying the proton calculations by a scaling factor, which was found to be 1.4 [10].

The flux of primary GCR particles varies over the time. Solar modulation is the dominant source of the observed variability. Therefore, we took the primary GCR spectrum in the form that accounts for this influence, with a modulation parameter, Φ [16]. We have used this expression elsewhere [10] for calculations involving GCR particles. In these calculations, the model Toluca meteoroid was irradiated by a homogeneous and isotropic GCR flux. We used only one value of the modulation parameter, $\Phi = 550$ MeV, which is very close to a long-term average [17]. The effective flux of protons above 10 MeV was 4.8 protons $cm^{-2}\ s^{-1}$. This effective flux was determined from the fitting of Knyahinya experimental data for cosmogenic radionuclides [11].

For these calculations, Toluca was modeled as a 3.9 m radius sphere with the composition of a typical IA iron meteorite (91% Fe, 8% Ni, 0.5% Co, 0.3% P and 0.1% C). We used a density of 7.8 g cm^{-3}. To examine the depth dependence of particle fluxes, the sphere was divided into spherical shells with thicknesses of 4 cm. The fluxes of neutrons and corresponding capture rates were calculated for each cell.

Statistical errors of neutron fluxes calculated using this geometric model and running 100,000 primary GCR protons were at the level of 3–5%. The systematic uncertainties of our calculated fluxes are not known but are probably of the order of 10%.

The capture rate of neutrons on isotope i (g^{-1} s^{-1}) at a depth D in a model sphere with a radius R is:

$$P_i(R,D) = N_i \int_0^\infty \sigma_i(E) J(E,R,D) \mathrm{d}E \qquad (1)$$

where N_i is the number of atoms for target isotope i per gram material in the sample, σ_i (cm^2) is the cross section for the capture of neutrons with energy E on the isotope i, and $J(E,R,D)$ (cm^{-2} s^{-1} MeV^{-1}) is the flux of neutrons with energy E (MeV) at location D inside the irradiated body. As mentioned above, the secondary neutron fluxes $J(E,R,D)$ for GCR particles are calculated using LCS.

The rates for the capture of neutrons by ^{182}W, ^{183}W, and ^{186}W were calculated using the detailed library of (n,γ) cross sections in MCNP. These W isotopes have thermal cross sections and resonance integrals of (20.7 and 604 barns), (10.1 and 337 b), and (37.9 and 485 b), respectively [18], but these cross sections are only crude guides to relative rates for $W(n,\gamma)$ reactions because the detailed structure of resonances and of the calculated fluxes of low-energy neutrons can significantly affect the MCNP-calculated rates. The $^{184}W(n,\gamma)$ cross sections are much lower, and thus we did not calculate rates for that reaction. The uncertainties in the calculated rates are hard to quantify but are believed to be less than 30%.

3. Results and discussion

The principal feature of the depth dependence of the thermal plus epithermal neutron flux is a maximum at a depth of 40 cm and decreases exponentially at greater depth with an e-folding length of about 30–34 cm. This neutron-flux profile is similar to those obtained in earlier calculations [12,13,15].

Having calculated the fluxes of thermal and epithermal neutrons, we calculated capture rates on various tungsten isotopes using Eq. (1). The calculated rate versus depth profiles for the neutron-capture reactions on tungsten are presented in Fig. 1.

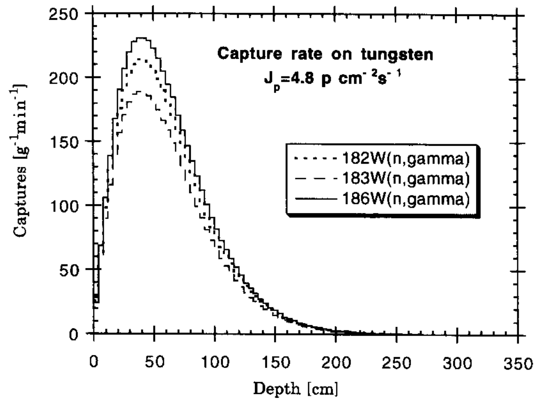

Fig. 1. Calculated capture rates on tungsten isotopes in the Toluca meteorite. The results are normalized to primary GCR-particle flux of 4.8 nucleons cm^{-2} s^{-1}.

Depth profiles of all these isotopes have very similar shapes, with their peak rates at the depth of about 40 cm being about 7.8 times those for 0–4 cm. The calculated capture rates then drop with increasing depth.

For this study of the possible effect of $W(n,\gamma)$ reactions on W isotope systematics, we consider the peak rates which are 213, 194, and 237 captures min^{-1} (g-isotope)$^{-1}$ for $W(n,\gamma)$ reactions with ^{182}W, ^{183}W, and ^{186}W, respectively. The relative burnout, r_i, of the particular isotope i is given by:

$$r_i = P_i T_{ex} / n_i \qquad (2)$$

where P_i is the rate for neutron-capture reaction on isotope i, T_{ex} is exposure age of investigated body, and n_i is the number of a particular tungsten isotope i atoms in 1 g. Using LCS-calculated capture rates

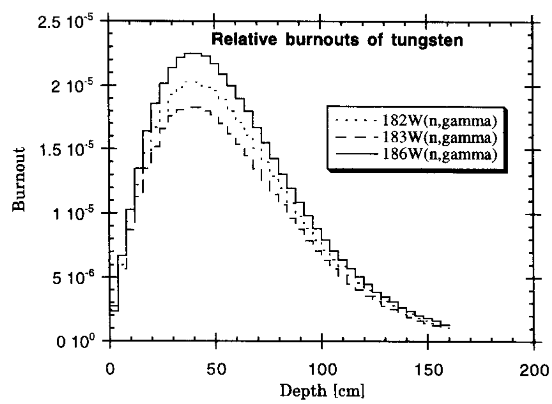

Fig. 2. Relative burn-out of tungsten isotopes in Toluca meteorite using LCS-calculated capture rates and 600 Ma exposure.

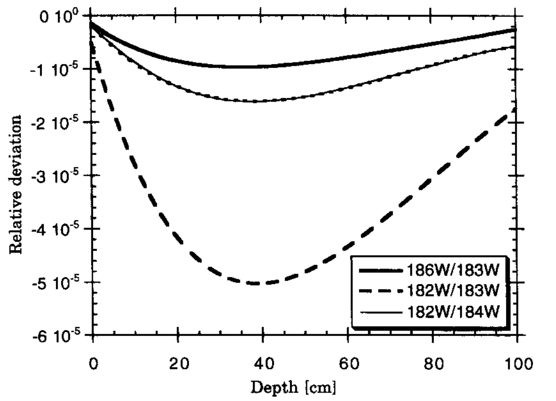

Fig. 3. Relative deviations of normalized tungsten isotopic ratios for Toluca meteorite. $^{182}W/^{183}W$ and $^{186}W/^{183}W$ are normalized to $^{184}W/^{183}W$ and $^{182}W/^{184}W$ is normalized to $^{186}W/^{184}W$.

and a 600 Ma exposure [19], the relative depth-dependent burn-out rates (Fig. 2) were calculated. Maximum burn-out rates were found to be 2.0×10^{-5}, 1.85×10^{-5}, and 2.3×10^{-5} for ^{182}W, ^{183}W, and ^{186}W, respectively. The relative deviations from standard values for various tungsten isotopic ratios were calculated using the formula:

$$\epsilon = \left[({}^{i}W/{}^{j}W)_{calc} - ({}^{i}W/{}^{j}W)_{std} \right] / ({}^{i}W/{}^{j}W)_{std} \quad (3)$$

For the comparison with experiments [1], the calculated values for $^{182}W/^{183}W$, are normalized to the $^{184}W/^{183}W$ ratio. Resulted deviations are shown in Fig. 3. After this correction, the maximum rates would cause decreases in the normalized $^{182}W/^{183}W$ and $^{186}W/^{183}W$ ratios of about 5 and 1 parts in 10^5, respectively.

While Toluca was the first meteorite analyzed for its W isotopic composition, precise measurements have now been made on more than 40 different meteorites and lunar samples by the Michigan Group. The results of these new measurements are now expressed as deviations in $^{182}W/^{184}W$ normalized to $^{186}W/^{184}W$. This ratio for the Toluca meteorite is also given in Fig. 3.

Thus the calculated maximum change in the normalized $^{182}W/^{183}W$ ratio due to neutron-capture reactions (corresponding to the maximum in calculated capture rates) cannot account for more than ~ 25% of the mass-182 deficit observed in Toluca W by Harper and Jacobsen [1], which is therefore probably due to the decay of ^{182}Hf in the early solar system.

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